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Antiferromagnetism in Fe-doped $PrRh_2Si_2$: a Mössbauer study

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Abstract $PrRh_2Si_2$ is highly anisotropic Ising-type antiferromagnetic system. The ordering temperature ($T_N \sim 68$ K) of $PrRh_2Si_2$ is exceptionally high on the de-Gennes scale in the family of RRh_2Si_2 (R = rare earths). The reason for this anomalous behaviour is not clear. It is believed that the presence of uniaxial anisotropy assists in enhancing the T_N. The Mössbauer study was performed on a 10% Fe-doped $PrRh_2Si_2$ sample to understand the magnetic coupling between different sites of $PrRh_2Si_2$. The Mössbauer spectra together with the magnetic susceptibility data suggest that the magnetic coupling in $PrRh_2Si_2$ is provided mainly by the RKKY interaction between Pr-moments.

Keywords Antiferromagnetism · Magnetocrystalline anisotropy · Mössbauer spectra

1 Introduction

The RRh_2Si_2 (R = rare-earths) compounds form in $ThCr_2Si_2$-type body centered tetragonal structure (space group I4/mmm) in which rare-earth ions occupy the crystallographic site of tetragonal point symmetry. This type of structure is favourable for a variety of magnetic ground states and presents very rich physics. As for example, the antiferromagnetically ordered system $YbRh_2Si_2$ ($T_N \sim 70$ mK) exhibits heavy-fermion behaviour and non-Fermi liquid state, and is situated close to the quantum critical point in which the ordering temperature can be easily tuned either by the magnetic field or by the chemical pressure [1–4]. Another system $CeRh_2Si_2$ has

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very high antiferromagnetic ordering temperature ($T_N \sim 36$ K) and exhibits pressure induced superconductivity [5–9]. On the basis of de-Gennes scaling in the series of RRh_2Si_2 one expects an ordering temperature of only 1.2 K in $CeRh_2Si_2$. Similarly the magnetic susceptibility of $PrRh_2Si_2$ exhibits an antiferromagnetic ordering at 68 K, which is again anomalously high compared to the de-Gennes expected transition temperature of 5.4 K in this compound [10]. In spite of numerous investigations made on $CeRh_2Si_2$, the precise nature of magnetism i.e. localized moment vs. itinerant moment characteristics could not be settled. The Pr-based systems are especially interesting due to the prominent role of crystal electric field effect in determining the ground state. The present investigation is aimed to understand better the magnetic coupling between different sites of $PrRh_2Si_2$ as well as the nature of antiferromagnetism in this system. We present here our results based on magnetic susceptibility measurement and Mössbauer study of 10% Fe-doped $PrRh_2Si_2$.

2 Experimental

The polycrystalline sample of Fe-doped $PrRh_2Si_2$ was prepared by the standard arc melting on a water cooled copper hearth in an Argon atmosphere and subsequent annealing at 1,000°C for a week. The sample was characterized by the powder X-ray diffraction and scanning electron micrographs. The magnetic susceptibility was measured using a commercial SQUID magnetometer. Mössbauer spectra were recorded at room temperature in a transmission geometry using a conventional ^{57}Fe constant acceleration Mössbauer spectrometer employing a 25 mCi ^{57}Co/Rh source. The spectra were analyzed using least squares method assuming Lorenzian lineshapes. The isomer shift and quadrupole splitting (QS) have an uncertainty of about 0.02 mm/s and the hyperfine field (B_{hf}) has an uncertainty of about 0.1 T. Relative areas are accurate up to 2%.

3 Results and discussion

The powder X-ray diffraction shows that $Pr(Rh_{0.9}Fe_{0.1})_2Si_2$ forms in single phase in $ThCr_2Si_2$-type tetragonal structure (I4/mmm) with lattice parameters $a = 4.071$ Å, $c = 10.112$ Å and unit cell volume = 167.57 Å^3. The unit cell volume of Fe-doped system is slightly less compared to that of undoped-$PrRh_2Si_2$ ($a = 4.077$ Å, $c = 10.148$ Å and unit cell volume = 168.67 Å^3) which is in accordance with different atomic radii of Rh and Fe.

Figure 1 shows the temperature dependence of magnetic susceptibility data of doped- and undoped-$PrRh_2Si_2$. While undoped-$PrRh_2Si_2$ orders antiferromagnetically below 68 K the Fe-doped one orders at 52 K. Thus 10% Fe doping reduces the ordering temperature from 68 to 52 K. This behaviour is consistent with the reported antiferromagnetic transition temperature of 7.7 K in $PrFe_2Si_2$ (complete replacement of Rh by Fe) [11]. In paramagnetic state the magnetic susceptibility of both the doped- and undoped-$PrRh_2Si_2$ follows the Curie–Weiss behaviour. For the case of doped-$PrRh_2Si_2$ we got an effective moment of 3.84 μ_B and θ_p of +2.08 K

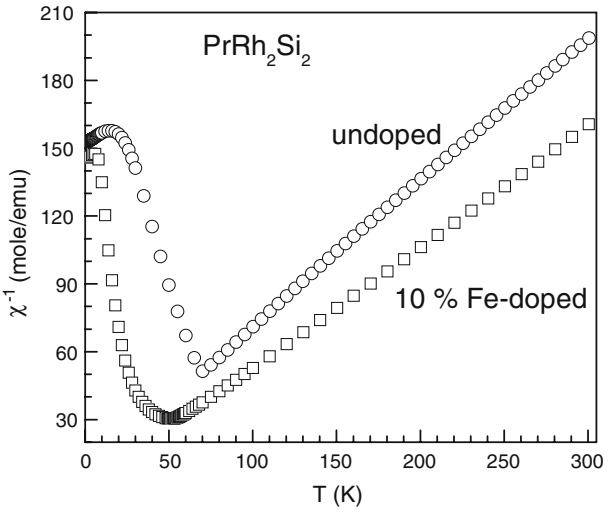

Fig. 1 The temperature dependence of inverse magnetic susceptibility data of Fe-doped and undoped-$PrRh_2Si_2$ measured at 1 T

from the linear fit of inverse magnetic susceptibility data in the temperature range of 100–300 K which differ from the values of $\mu_{eff} = 3.55\ \mu_B$ and $\theta_p = -13.96$ K for undoped-$PrRh_2Si_2$. The theoretically expected effective moment for Pr^{3+} ions is 3.58 μ_B. The Curie–Weiss temperature changes from a negative value in undoped-$PrRh_2Si_2$ to a positive value in doped-$PrRh_2Si_2$ indicating a dominant ferromagnetic exchange in the doped one in contrast to an antiferromagnetic exchange in the undoped-$PrRh_2Si_2$.

^{57}Fe Mössbauer spectra of Fe-doped $PrRh_2Si_2$ sample were recorded at room temperature (far above the T_N) as well as at 18 K (well below the T_N) which are shown in Fig. 2. It is evident from the figure that the spectra consist of a single absorption line which is well represented by Lorentzian curve without any hyperfine or quadrupolar splitting in both the cases. No noticeable change is observed in the nature of the spectra at room temperature and 18 K except that the spectrum has broadened at 18 K. The Mössbauer spectrum of $PrFe_2Si_2$ is also reported to exhibit no quadrupolar and hyperfine splitting and is represented by a single Lorentzian curve [11]. For the spectrum recorded at room temperature (Fig. 2) the isomer shift was found to be 0.24 mm/s and a linewidth of 0.34 mm/s which is slightly larger than that expected from a well crystalline single line spectrum in thin absorber approximation (0.25 mm/s). Whereas for the spectrum recorded at 18 K (Fig. 2) the isomer shift is 0.37 mm/s and linewidth = 0.63 mm/s. This line broadening may be attributed to unresolved quadrupolar splitting or slight vibration effect from the closed cycle helium cryostat. The absence of hyperfine splitting clearly suggests that there is no coupling between the nuclear magnetic moment of Fe with hyperfine magnetic field at the Fe site. Since we observe an antiferromagnetic ordering in doped-$PrRh_2Si_2$ in magnetic susceptibility measurement, but no splitting in Mössbauer spectra, this clearly means that Fe sites and hence Rh sites do not carry appreciable internal magnetic field and the magnetic ordering comes mainly from Pr-Pr coupling. Further, since the Rh atoms seem not to carry magnetic moment, we speculate the nature of magnetism in $PrRh_2Si_2$ to be localized.

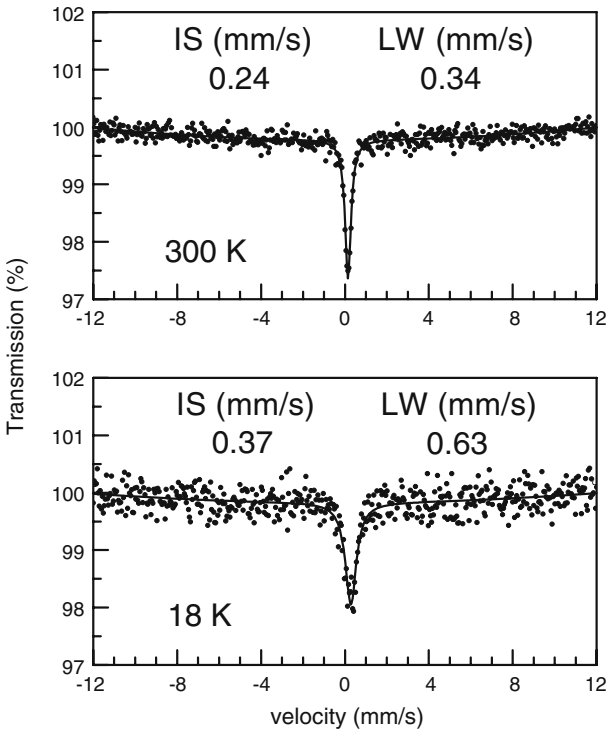

Fig. 2 ^{57}Fe-Mössbauer spectra of Fe-doped $PrRh_2Si_2$ at room temperature and 18 K. The *solid lines* represent the best fit to the data according to the Lorentzian fit

References

1. Trovarelli, O., Geibel, C., Mederle, S., Langhammer, C., Grosche, F.M., Gegenwart, P., Lang, M., Sparn, G., Steglich, F.: $YbRh_2Si_2$: Pronounced non-Fermi-liquid effects above a low lying magnetic phase transition. Phys. Rev. Lett. **85**, 626 (2000)
2. Gegenwart, P., Custers, J., Geibel, C., Neumaier, K., Tayama, T., Tenya, K., Trovarelli, O., Steglich, F.: Magnetic-field induced quantum critical point in $YbRh_2Si_2$. Phys. Rev. Lett. **89**, 056402 (2002)
3. Plessel, J., Abd-Elmeguid, M.M., Sanchez, J.P., Knebel, G., Geibel, C., Trovarelli, O., Steglich, F.: Unusual behaviour of the low-moment magnetic ground state of $YbRh_2Si_2$ under high pressure. Phys. Rev. B **67**, 180403(R) (2003)
4. Custers, J., Gegenwart, P., Wilhelm, H., Neumaier, K., Tokiwa, Y., Trovarelli, O., Geibel, C., Steglich, F., Pépin, C., Coleman, P.: The break-up of a heavy electrons at a quantum critical point. Nature **424**, 524 (2003)
5. Graf, T., Hundley, M.F., Modler, R., Movshovich, R., Thompson, J.D., Mandrus, D., Fisher, R.A., Phillips, N.E.: Magnetic phase transitions in $CeRh_2Si_2$: specific heat, susceptibility, and resistance studies. Phys. Rev. B **57**, 7442 (1998)
6. Kawarazaki, S., Sato, M., Miyako, Y., Chigusa, N., Watanabe, K., Metoki, N., Koike, Y., Nishi, M.: Ground-state magnetic structure of $CeRh_2Si_2$ and the response to hydrostatic pressure as studied by neutron diffraction. Phys. Rev. B **61**, 4167 (2000)
7. Gómez Berisso, M., Pedrazzini, P., Sereni, J.G., Trovarelli, O., Geibel, C., Steglich, F.: Study of $Ce(Rh_{1-x}Pd_x)_2Si_2$: pronounced differences between $CeRh_2Si_2$ and $CePd_2Si_2$ ground states. Eur. Phys. J. B. **30**, 343 (2002)

8. Araki, S., Settai, R., Kobayashi, T.C., Harima, H., Ōnuki, Y.: Fermi surface instability in $CeRh_2Si_2$ under pressure. Phys. Rev. B **64**, 224417 (2001)
9. Araki, S., Nakashima, M., Settai, R., Kobayashi, T.C., Ōnuki, Y.: Pressure-induced superconductivity in an antiferromagnet $CeRh_2Si_2$. J. Phys., Condens. Matter **14**, L377 (2002)
10. Anand, V.K., Hossain, Z., Behr, G., Chen, G., Nicklas, M., Geibel, C.: Magnetocrystalline anisotropy and antiferromagnetic phase transition in $PrRh_2Si_2$. J. Phys., Condens. Matter **19**, 506205 (2007)
11. Malaman, B., Venturini, G., Blaise, A., Sanchez, J.P., Amoretti, G.: Magnetic study of $PrFe_2Si_2$ and $PrFe_2Ge_2$ by susceptibility measurements, neutron diffraction and Mössbauer spectroscopy. Phys. Rev. B **47**, 8681 (1993)